



# Implications of Industrial Loads for Ammonia Pollution in an Urban Lake

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**ABSTRACT:** The recent history of loading of total ammonia (T-NH<sub>3</sub>) and organic nitrogen (N) from a pharmaceutical manufacturing facility to a municipal treatment plant (Metro) in Syracuse, New York, and the discharge of these constituents from Metro to N-polluted Onondaga Lake is documented. Further, the benefit of the implementation of pretreatment at the pharmaceutical plant, and the effect of an upset event at this treatment facility on loading to Metro and the lake and in-lake concentrations are also documented. Models are used as analytical tools to couple loading and in-lake concentrations, to delineate the role that this pharmaceutical facility has played in the lake's ammonia pollution problem, and to evaluate the potential implications of future pretreatment upset events for the success of a rehabilitation program that is underway for the lake. The responsiveness of the lake to reductions in external loading is established by the lower T-NH<sub>3</sub> concentration observed in the upper waters of the lake in the spring of 1999. Model analysis demonstrates this reduction was primarily (~ 75%) because of the decrease in loading from the pharmaceutical facility achieved by pretreatment. An abrupt increase in loading in May 1999 associated with an upset event at the pretreatment facility caused a corresponding increase in the T-NH<sub>3</sub> concentration of the lake of approximately 0.5 mg N/L. Model projections demonstrate that the load from the pharmaceutical plant before construction of the pretreatment facility exacerbated the lake's ammonia problems by increasing the occurrence and margin of violations of the toxicity standard. Continued upset events at the pretreatment plant could compromise the lake rehabilitation program. *Water Environ. Res.*, 73, 192 (2001).

**KEYWORDS:** ammonia, industrial pretreatment, lakes, pharmaceutical waste, models.

## Introduction

Industrial discharges can represent substantial portions of the waste loads received by municipal wastewater treatment plants (WWTPs) (Droste, 1997, and Metcalf and Eddy, 1991). These industrial inputs can be highly variable because they are associated with manufacturing schedules and irregularities in the performance of pretreatment facilities (Metcalf and Eddy, 1991). Municipal facilities often have limited capacities to treat pulse-like industrial loads (Droste, 1997). Such loading events may be transmitted, at least partially, to surface waters that receive effluent from affected WWTPs. Receiving water in urban areas often has limited assimilative capacity to accommodate abrupt increases in pollutant loadings (Chapra, 1997, and Thomann and Mueller, 1987). Loading events are particularly problematic for lakes, as the interval of effect can be lengthy (e.g., compared with streams and rivers) because of slower flushing rates (Chapra, 1997).

This paper documents the recent history of loading of total ammonia (T-NH<sub>3</sub>) and organic nitrogen (N) from a pharmaceutical manufacturing facility to a WWTP in Syracuse, New York, and the discharge of these constituents to N-polluted Onondaga Lake (Brooks and Effler, 1990; Effler et al., 1990; and Matthews et al.,

2000). The benefit of performing pretreatment at the pharmaceutical plant and the effect of an upset event at this facility, related to the lake's status with respect to ammonia toxicity criteria (U.S. EPA, 1985 and 1998), are documented. Empirical models and a mass-balance N model for the lake are tested against the conspicuous signatures of T-NH<sub>3</sub> concentrations imparted to the upper water by pretreatment and the upset event. A probabilistic modeling framework that incorporates the mass-balance N model (Gelda et al., 2001) is applied to evaluate the role that this pharmaceutical facility has played in the lake's ammonia pollution problem and the implications that future upset events could have on the success of the rehabilitation program that is underway for the lake.

## Study System

**Municipal Wastewater Treatment Plant.** The Metropolitan Syracuse Wastewater Treatment Plant (Metro) serves approximately 300 000 residents of Onondaga County, New York (including the City of Syracuse) and a number of local industries. Effluent from this facility is discharged to the southern end of Onondaga Lake (Figure 1) via a surface outfall. The existing facility was designed to treat an average flow of 3.5 m<sup>3</sup>/s (80 mgd); flows as great as 5.3 m<sup>3</sup>/s (120 mgd) receive full (tertiary) treatment. Greater flows (e.g., runoff events) receive only partial treatment before discharge to the lake. The Metro presently represents approximately 20% of the annual inflow to the lake and often is the single largest source of water during the low fluvial flow interval of late summer (Effler et al., 1996a).

The Metro was not designed to achieve significant nitrification (contact stabilization modification of activated sludge; U.S. EPA, 1993). However, substantial nitrification has been attained within the facility during the warmer months of certain years since the mid-1980s (Effler et al., 1996a). This causes seasonal shifts in the contributions of T-NH<sub>3</sub> and oxidized forms of N (nitrate (NO<sub>3</sub><sup>-</sup>), nitrite (NO<sub>2</sub><sup>-</sup>), and the sum of NO<sub>3</sub><sup>-</sup> and NO<sub>2</sub><sup>-</sup> : NO<sub>x</sub><sup>-</sup>) to the facility's N load (Effler et al., 1996a, and Gelda et al., 1999, 2000, and 2001). The Metro is the dominant source of N and phosphorus (P) for Onondaga Lake, presently representing 80, 90, and 60% of the total annual loads of total N (TN), T-NH<sub>3</sub>, and P, respectively (Effler et al., 1996a). The prevailing annual areal loads of N (~ 200 g/m<sup>2</sup>·a) and P (~ 8 g/m<sup>2</sup>·a) (Effler et al., 1996a) are among the highest reported in the literature (Brezonik, 1972, and Stauffer, 1985).

**Pharmaceutical Waste, Pretreatment.** A pharmaceutical manufacturing facility, owned and operated by Bristol-Myers Squibb Company (BMS), is located in the metropolitan Syracuse area. This facility has been primarily engaged in the production of bulk antibiotics. Process wastewater was initially discharged directly to a tributary of Onondaga Lake. Later this wastewater was treated at a smaller (~ 0.9 m<sup>3</sup>/s [20 mgd]) regional municipal treatment

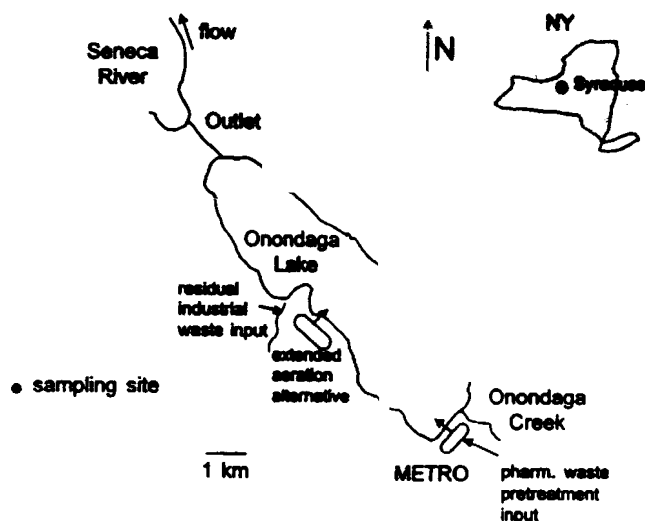


Figure 1—Onondaga Lake setting; discharges, lake sampling site, and river.

facility. Subsequently (~ 1980) it was delivered to Metro for treatment. Pretreatment of this industry's wastewater was mandated in 1992 and began in late 1996.

This industry's wastewater is enriched in solids, total Kjeldahl nitrogen (TKN), T-NH<sub>3</sub>, P, sulfate, phenol, and several organic solvents (Engineering Science, 1994). The characterization of TKN loading from this discharge that prevailed before pretreatment (Table 1) is based on monitoring conducted over a 7-month period in 1992 (Engineering Science, 1994). The average discharge flow and TKN concentration for the study interval were  $56.4 \times 10^{-3} \text{ m}^3/\text{s}$  and 588 mg N/L, respectively, and the average TKN load was approximately 2900 kg/d (Table 1). The concentrations and loads were more variable than the discharge flow (Table 1). The measurements of T-NH<sub>3</sub> were much more limited, the average for three samplings was 218 mg N/L (Engineering Science, 1992). Later a T-NH<sub>3</sub> concentration of 250 mg N/L was assumed in characterizing this industrial waste stream (Engineering Science, 1994). Provisional pretreatment limits require a reduction in the TKN load of 93%; a 90% reduction in the T-NH<sub>3</sub> load is required if the assumed T-NH<sub>3</sub> concentration is accepted. The treatment scheme for this industrial waste initially included primary pretreatment using low-rate anaerobic reactors, followed by aerobic polishing using sequencing batch reactors. The features of process design and operation of this facility continue to evolve in an effort to improve performance.

**Onondaga Lake and Tributaries.** Onondaga Lake is a hard-water, alkaline, dimictic system, located in metropolitan Syracuse, New York (Figure 1). This lake has a volume of  $131 \times 10^6 \text{ m}^3$ , a surface area of 12.0 km<sup>2</sup>, and a maximum depth of 19.5 m (Effler, 1996). The watershed supports a population of approximately 450 000. The lake discharges through a single outlet at its northern end to the Seneca River. Onondaga Lake was oligo-mesotrophic before European settlement in the late 1700s (Rowell, 1996), and supported a cold-water fish population (Tango and Ringler, 1996). Reception of increasing loads of domestic and industrial waste from the watershed accompanied development and urbanization and led to profound degradation of the lake and the loss of the use of its original resources (Effler, 1996). The lake is now hypereutrophic and severely polluted as a result of waste inputs (Effler

and Harnett, 1996; Effler and Hennigan, 1996; and Hennigan, 1991). Manifestations of hypereutrophy, driven by P inputs from Metro, include frequent and severe phytoplankton blooms, poor clarity, and extremely degraded oxygen resources (Address and Effler, 1996; Effler et al., 1988 and 1996a; and Gelda and Auer, 1996).

Concentrations of several forms of N are extremely high in Onondaga Lake because of the inputs from Metro (Brooks and Effler, 1990; Canale et al., 1996; and Gelda et al., 1999). Concentrations of T-NH<sub>3</sub> increase (caused by ammonification and sediment release) and NO<sub>x</sub> is depleted (denitrification) in the anoxic-anaerobic hypolimnion during summer stratification (Brooks and Effler, 1990). This paper focuses on the upper waters of the lake because the lack of oxygen in the hypolimnion limits the vertical distribution of the lake's fishes (Tango and Ringler, 1996). Water quality criteria (U.S. EPA, 1998) and state standards for ammonia (Matthews et al., 2000) and NO<sub>2</sub><sup>-</sup> (Gelda et al., 1999) are exceeded or violated for extended intervals and by wide margins in the upper waters of the lake. Concentrations of T-NH<sub>3</sub> and NO<sub>3</sub><sup>-</sup> (sum > 2 mg N/L) in these layers remain well above levels considered limiting to phytoplankton growth (Canale et al., 1996).

Matthews et al. (2000) recently documented temporal patterns of T-NH<sub>3</sub> in the upper waters of Onondaga Lake, and status with respect to ammonia toxicity criteria, for the spring to fall interval of 10 consecutive years (1989 to 1998). Substantial interannual and seasonal variations were reported for T-NH<sub>3</sub> concentrations. Empirical analyses indicated that the wide interannual differences in concentrations reported at spring turnover (1.5 to 3.5 mg N/L) were driven largely by natural variations in antecedent tributary flow. Interannual differences in the subsequent patterns over the spring-to-fall interval were attributed to year-to-year differences in the relative magnitudes of the sources and sinks of T-NH<sub>3</sub>; primarily these were the extent of nitrification achieved within Metro during summer and the irregular occurrence of nitrification events in the lake during fall mixing (Gelda et al., 2000).

The hydrology of the lake is well quantified; more than 90% of the surface inflow (including Metro) has been continuously gauged since the early 1970s (Effler and Whitehead, 1996). Direct ground-water inputs are insignificant and fluxes associated with direct precipitation and evaporation are approximately in balance on an annual basis (Effler and Whitehead, 1996). Strong seasonal and interannual variations in hydrologic loading to the lake occur;

Table 1—Nitrogen loading from pharmaceutical facility (BMS) before pretreatment and proposed pretreatment limits (Engineering Science, 1994).

Parameter	Before treatment			Pretreatment limits
	Average	CV <sup>a</sup>	n <sup>b</sup>	
	56.4	0.13	197	57
	588	0.24	76	40
	2887	0.24	76	197
	250 <sup>c</sup>	—	—	25
	1230	—	—	123

<sup>a</sup> CV: coefficient of variation.

<sup>b</sup> n: number of observations

<sup>c</sup> Assumed.

typically flows are greatest in March and April and lowest in the July to September interval. The lake flushes rather rapidly with an average of 4 (completely mixed) flushes per year (Gelda et al., 2001) and thus responds quickly to abrupt changes in material loading (Chapra, 1997). Tributary loading of N, particularly as T-NH<sub>3</sub>, is minor (~ 10%) relative to Metro (Effler and Whitehead, 1996).

**Rehabilitation Program.** Onondaga County has committed to a significant rehabilitation program to eliminate water quality violations in Onondaga Lake associated with the discharge of municipal wastewater. The presently approved program, to be phased over 15 years (3 phases), is the largest public works project ever undertaken by the community (≥ U.S.\$380 000 000). Much of this program is associated with the upgrade of Metro to greatly reduce P and T-NH<sub>3</sub> discharges. The first phase limits T-NH<sub>3</sub> and total P concentrations (averages) in the Metro effluent to 1997 levels. The aeration system for biological treatment was upgraded in 1998. The effluent concentration limits for T-NH<sub>3</sub> for phase 2 (starting in 2003) would be 1.65 and 3.3 mg N/L for the June-to-October and November-to-May intervals, respectively. These limits would be reduced to 1.0 and 2.0 mg N/L, respectively, by 2013 to meet phase 3 requirements. The estimated cost for the nitrification treatment at Metro is U.S.\$130 000 000. Nitrification would be achieved with biological aerated filters, where active biomass is attached to submerged granular media (U.S. EPA, 1993). Effective pretreatment of the waste generated by BMS is considered to be an important component for meeting the specified effluent goals. An alternate plan for municipal wastewater treatment has been proposed that would achieve a maximum effluent T-NH<sub>3</sub> concentration of 1.0 mg N/L year-round, through the process of extended aeration (U.S. EPA, 1993; Figure 1). This greater level of treatment may be necessary to meet water quality standards (Gelda et al. 2001), with a reasonable margin of safety (U.S. EPA, 1991b).

## Methods

**Input Concentrations and Loads.** Estimates of mass loading rates of T-NH<sub>3</sub>, NO<sub>x</sub>, and organic N (TKN minus T-NH<sub>3</sub>) to Metro and Onondaga Lake were based on flow and concentration data collected by four different monitoring programs: (1) tributary flow measurements made by the U.S. Geological Survey (USGS), Ithaca, New York, (2) tributary concentrations measured by the Upstate Freshwater Institute (Syracuse) over several years (Effler and Whitehead, 1996), (3) influent and effluent flows and concentrations measured at Metro as part of the facility's permit requirements, and (4) effluent flow and concentrations measured at the BMS treatment facility as part of its permit requirements. Concentrations of these forms of N were determined according to standard methods (APHA et al., 1992, and U.S. EPA, 1983). Tributary samples were grab type and influent and effluent samples were flow-weighted 24-hour composites. At Metro, concentrations of T-NH<sub>3</sub>, NO<sub>x</sub>, and TKN were measured 5, 2, and 2 days per week respectively. At the BMS facility, T-NH<sub>3</sub> and TKN were determined approximately 7 days per month. These observations were augmented at times by measurements made by the staff of Metro. Effluent loads and the influent load for Metro were estimated as the product of daily flows and daily concentrations. Effluent concentrations for days without measurements were estimated by time interpolation (Canale et al., 1996, and Effler et al., 1996a).

Daily flows for the gauged tributaries for the 1988 to September 1998 interval were those reported by USGS. Flows for ungauged tributaries were estimated according to the protocol of Gelda et al.

(2001). Tributary flows for the October 1998 to June 1999 interval were based on data provided by USGS for Onondaga Creek, one of the two largest tributaries, for which the accuracy of measurements is rated as good (95% of daily discharge values within 10% of true value). Flow in this tributary is a good surrogate measure of the discharge from the other fluvial inputs (Effler and Whitehead, 1996, and Gelda et al., 2001). Daily loads were estimated as the product of the daily flows and constituent concentrations. These concentrations were specified by flow-concentration relationships for each of the tributaries, which represent prevailing conditions (Effler and Whitehead, 1996, and Gelda et al., 2001). The uncertainty in tributary loads associated with these protocols does not have a substantial effect on the model analyses presented here because of the dominance of Metro in regulating total loads to the lake (Gelda et al., 2001).

**Lake Monitoring and Calculations of Status for Ammonia Toxicity Standard.** The lake was monitored weekly (at mid-morning) during the April to early June interval of 1999 at a buoyed deep water (~ 19.5 m) location (Figure 1) in the lake's southern basin, which was found to be representative of lake-wide conditions (Effler, 1996). Field measurements of temperature (T) and pH were made at 1-m depth intervals with a Hydrolab Surveyor 3 (Austin, Texas), calibrated according to the manufacturer's instructions. Lake samples for laboratory analyses of T-NH<sub>3</sub> (U.S. EPA, 1983), NO<sub>x</sub> (U.S. EPA, 1983), TN (Valderrama, 1981), and chlorophyll a (Parsons et al., 1984) were collected, at the time of instrumental profiles, at depth intervals of 2 m. The concentration of organic N in the lake is calculated as the residual of TN and the sum of T-NH<sub>3</sub> and NO<sub>x</sub>. Chlorophyll a is included as a measure of phytoplankton biomass to support estimates of net phytoplankton growth (Canale et al., 1996).

There have been five revisions of the national criteria (chronic and acute, for salmonid and nonsalmonid receiving waters) to protect against the potential toxic effects of ammonia over the past 15 years (Heber and Ballentine, 1992, and U.S. EPA, 1985, 1996, 1998, and 1999). The changes have recently been reviewed by Matthews et al. (2000). The status of the lake over the April to June interval of 1999 is evaluated with respect to the present New York State standard for ammonia (which coincides numerically with the U.S. EPA's 1985 chronic criterion for nonsalmonid waters, the criterion continuous concentration [CCC]) by comparing the observed T-NH<sub>3</sub> concentrations with the standard. The value of the standard (or CCC) is a function of pH and T, reflecting the dependencies of the partitioning between the ammonium ion (NH<sub>4</sub><sup>+</sup>) and free ammonia (NH<sub>3</sub>, the more toxic form) and the toxicity of these conditions (U.S. EPA, 1999). The value of the standard decreases (i.e., more stringent limit) as pH (in particular) and T increase. Projections with the probabilistic framework are evaluated within the context of the standard and a recent revision of the national criterion (U.S. EPA, 1998).

**Nitrogen Model, Probabilistic Framework, and Applications.** A mechanistic dynamic mass-balance model for the principal forms of N, including T-NH<sub>3</sub>, NO<sub>x</sub>, particulate organic N (PON), and dissolved organic N (DON), has been developed and tested for Onondaga Lake (Canale et al., 1996). The water column of the lake is represented as two completely mixed vertical layers of fixed dimensions (demarcation depth of 8.5 m): an upper mixed layer (UML) and lower mixer layer (LML), corresponding approximately to the dimensions of the epilimnion and hypolimnion in summer (Effler and Owens, 1996). This representation is widely used in mass-balance models for stratifying lakes (Chapra, 1997,

and Thomann and Mueller, 1987). Pathways in the lake's N cycle represented in the model are: net phytoplankton growth, nitrification, denitrification, hydrolysis of DON, decomposition of PON, sediment release of T-NH<sub>3</sub>, settling of PON, vertical mixing-based exchange between the UML and LML, volatilization of NH<sub>3</sub>, external loading, and export from the basin (Canale et al., 1996). The preferential uptake of T-NH<sub>3</sub>, instead of NO<sub>x</sub>, by phytoplankton (for energetic reasons; Wetzel, 1983) is accommodated. The time series of net phytoplankton growth, which specifies the associated sink for T-NH<sub>3</sub>, was determined through model calibration procedures that resulted in predicted chlorophyll concentrations approximately matching observations (e.g., Canale et al., 1996). Independent determination of several model coefficients, based on field and laboratory experiments, enhanced the credibility of the model (Canale et al., 1996). The model was originally tested for the disparate forcing conditions and in-lake patterns of 2 years (Canale et al., 1996); subsequently this was extended to other years (Gelda et al., 2000). This testing has focused on the spring-to-fall interval.

A probabilistic modeling framework, which uses the N model, was developed to support *a priori* (futuristic) simulations of T-NH<sub>3</sub> and status with respect to the chronic criterion (Gelda et al., 2001), to guide management deliberations for related features of the rehabilitation program. The framework accommodates variability in forcing and ambient conditions, including: tributary runoff, rate of in-lake nitrification, treatment performance at Metro, lake pH, and lake temperature (Gelda et al., 2001). Variability in these conditions is specified through system-specific records of measurements and calculations that extend as long as 27 years (1971 to 1997). Sources of variability are represented in the probabilistic framework through both continuous long-term and Monte Carlo simulations (Gelda et al., 2001). Long-term data sets for tributary flow and vertical mixing are used to drive multiple-year simulations to quantify the variability caused by natural variations in these forcing conditions. Changes in material loading from tributaries associated with variations in flow are calculated based on prevailing flow-concentration relationships (Effler, 1996). The effects of a broad range of flow conditions (e.g., inclusion of runoff events) on nonpoint (tributary) T-NH<sub>3</sub> loading is well represented by the results of long-term monitoring of tributary concentrations (Effler, 1996), which were included in the development of the flow-concentration relationships (Gelda et al. 2001). Monte Carlo simulations accommodate the effects of interannual stochastic variations in nitrification in the lake (Gelda et al. 2000) and Metro and are used to calculate the ammonia criteria from distributions of pH and T formed from long-term measurements (Gelda et al., 2001). The framework performed well in representing the wide variations in T-NH<sub>3</sub> concentrations observed in the upper waters of the lake over a 6 year period. Predictions with the probabilistic framework can be presented in the form of frequency distributions of T-NH<sub>3</sub> concentrations and the exceedance count format (i.e., T-NH<sub>3</sub>/CCC > 1) of chronic criteria (U.S. EPA, 1991a and 1999).

Testing of the N model for the lake is extended in two ways here: (1) simulation of the spring turnover (April) T-NH<sub>3</sub> concentration annually over the 1988 to 1999 period, and (2) simulation of T-NH<sub>3</sub> concentrations in the UML over the early May to early June interval of 1999. In the first case, the simulation for each of the years is initialized by the concentrations measured during fall turnover of the previous year and is driven by Metro loading and tributary input information collected in each year over the October to April interval. These simulations test the hypothesis (Effler et

al., 1996a, and Matthews, et al. 2000) that the spring turnover T-NH<sub>3</sub> concentration is an effective integrator of the antecedent conditions because the model essentially treats T-NH<sub>3</sub> as a conservative substance over this interval because kinetic rates for the biochemical processes approach zero at low temperatures (Canale et al., 1996). Empirical models (Effler et al., 1996a) are also applied to resolve factors regulating the spring turnover concentration. Simulations with the N model for 1999, including extension to early June, are intended to quantitatively resolve the role variations in the industrial load played in imparting the conspicuous changes documented for T-NH<sub>3</sub> for that interval. Constituent concentrations for the UML for model state variables were calculated as volume-weighted values, based on the vertical profiles of measurements and hypsographic data (Doerr et al., 1996). Temporal distributions of vertical mixing for the April to June interval of 1999 were independently determined from T profiles, as described by Doerr et al. (1996). A value for the vertical mixing coefficient of 5 m/d is invoked (Doerr et al., 1996) for the months the lake is not monitored (November to March, late fall mixing through ice cover), which converts the two-layer framework to essentially a one-layer (i.e., well-mixed) system. The time step of loading inputs and model calculations is 1 day. Specifications for other model inputs and protocols have been described previously (Canale et al., 1996, and Gelda et al., 2000).

The probabilistic framework is applied to demonstrate the role that the industrial source of TKN has played in contributing to the observed levels of T-NH<sub>3</sub> in the lake and violations of the present toxicity standard and the implications of upset events at the industrial pretreatment facility, as characterized here for a single occurrence, for meeting the water quality goal for ammonia in the lake. Specifications of ambient conditions for the simulations were those described as appropriate by Gelda et al. (2001) for evaluation of management alternatives.

## Results and Discussion

**Concentrations and Loads of Nitrogen.** Influent loads of organic N and T-NH<sub>3</sub> to Metro demonstrated similar seasonality before pretreatment at BMS (somewhat lower levels in the July to August interval [Figures 2a and 2b]) that together imparted a more conspicuous seasonality in the TKN load (Figure 2c). The TKN load was approximately 30% lower in July and August than in most of the other months of the year. On average, T-NH<sub>3</sub> represented slightly more than half (53%) of the TKN load to Metro (Table 2). Assuming that the average TKN load reported for the BMS wastewater based on 1992 monitoring is representative, this industrial source contributed approximately one-third (an average of 2887 kg N/d) of the total load received by Metro (Tables 1 and 2). At the provisional permit loading limits (i.e., following pretreatment, Table 1), the BMS input to Metro would represent approximately 3% of the total loads of T-NH<sub>3</sub> and TKN received by this municipal facility.

The Metro removed approximately 80% of the organic N load over the 1989 to 1996 interval (Table 2), with only modest seasonality in performance (Figure 3a). The average concentration of organic N in the Metro effluent over the 1989 to 1996 interval was 3.2 mg N/L (Table 2). The monitoring data for the facility for this interval reaffirm, for a longer period, earlier observations (Effler et al., 1996a) that substantial treatment of T-NH<sub>3</sub> has been achieved during the warmer months, though significant interannual variations have occurred (Figure 3b). This seasonality reflects the strong dependence of the nitrification process on T (Figure 3d; U.S. EPA,

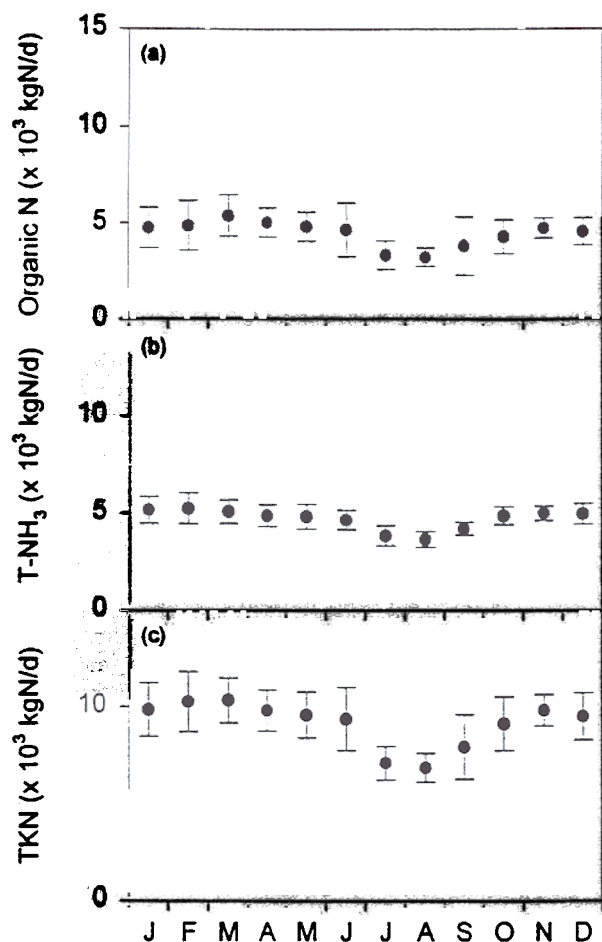


Figure 2—Monthly average ( $\pm$  standard deviation) loads of forms of N to Metro for 1989 to 1996 interval: (a) organic N, (b) T-NH<sub>3</sub>, and (c) TKN.

1993). On average, approximately 50% removals were attained in August, September, and October during the 1989 to 1996 interval (Figure 3b). Unusually high concentrations of NO<sub>2</sub><sup>-</sup> have been reported for the effluent during these months, indicating incomplete nitrification in the facility (Gelda et al., 1999). The seasonality and variability in the TKN load from Metro (Figure 3c) have

been largely driven by the temporal structure in the T-NH<sub>3</sub> load (Figure 3b). The average T-NH<sub>3</sub> concentration in the Metro effluent during the 1989 to 1996 interval was 13.6 mg N/L (Table 2). On average, during the 1989 to 1996 period, T-NH<sub>3</sub> was approximately 80% of the TKN load discharged to Onondaga Lake.

The time series of BMS effluent and Metro influent and effluent N concentrations and loads are presented in Figure 4 for the September 1998 to May 1999 interval to investigate the interplay between these conditions, and to support coupling of these forcing conditions to the subsequently reported lake concentrations of 1999. The effects of pretreatment clearly manifested in the September to April interval; organic N and T-NH<sub>3</sub> concentrations (Figure 4a) and loads (Figure 4b) were typically well below the averages reported for the discharge before commencement of pretreatment (Table 1). However, performance was highly variable over this interval; the proposed limits were exceeded for substantial portions of the interval, and by a rather wide margin, in mid-October, mid-January and mid- to late April (Figures 4a and 4b). Performance of the pretreatment facility was particularly good in November and December of 1998 (Figures 4a and 4b). The high concentrations (Figure 4a) and loads of N (Figure 4b), particularly T-NH<sub>3</sub>, in May of 1999, reflect a temporary failure (i.e., upset) at the pretreatment facility. Concentrations of TKN, mostly as T-NH<sub>3</sub>, increased progressively until late May (Figure 4a), to levels observed before construction of the pretreatment facility (Table 1). The May increase in loading from the pretreatment facility had the same general form, but it was somewhat diminished compared with the concentration time structure because of reduced discharge flows. The May T-NH<sub>3</sub> load from BMS (beyond the proposed permit limit) represented approximately 30% of the total load received by Metro in that month (Figure 4d), similar to the contribution from this industry before construction of the pretreatment facility.

It is important to establish the extent to which loads from BMS influenced the load from Metro; that is, the extent to which this industrial source may directly influence Onondaga Lake. Further, recent changes in performance of Metro in removing T-NH<sub>3</sub> (Figure 5) need to be identified and differentiated from the effects of pretreatment. Monthly loads of T-NH<sub>3</sub> from Metro to the lake during the June 1998 to February 1999 interval were the lowest observed to date (Figure 5), and are associated with the upgrade in the aeration system. Nearly complete treatment (Effler et al., 1996a) was achieved from July through mid-December of 1998 (i.e., effluent concentrations of T-NH<sub>3</sub> were typically < 1 mg N/L,

Table 2—Nitrogen loading to Metro, discharge from Metro, and percent removal at Metro: long-term averages for the 1989 to 1996 interval.

Nitrogen form	Influent			Effluent			Removal, %
	Average	CV <sup>a</sup>	n <sup>b</sup>	Average	CV	n	
Organic concentration, mg N/L	15.7	0.33	814	3.2	0.45	1005	
Organic load, kg/d	4336	0.38	814	864	0.54	1005	79.1
T-NH <sub>3</sub> concentration, mg N/L	17.6	0.25	2921	13.6	0.39	2922	
T-NH <sub>3</sub> load, kg/d	4716	0.21	2921	3470	0.39	2922	27.4
TKN concentration, mg N/L	33.0	0.24	816	16.8	0.33	1007	
TKN load, kg/d	9023	0.25	816	4324	0.36	1007	52.4

<sup>a</sup> CV: coefficient of variation.

<sup>b</sup> n: number of observations.

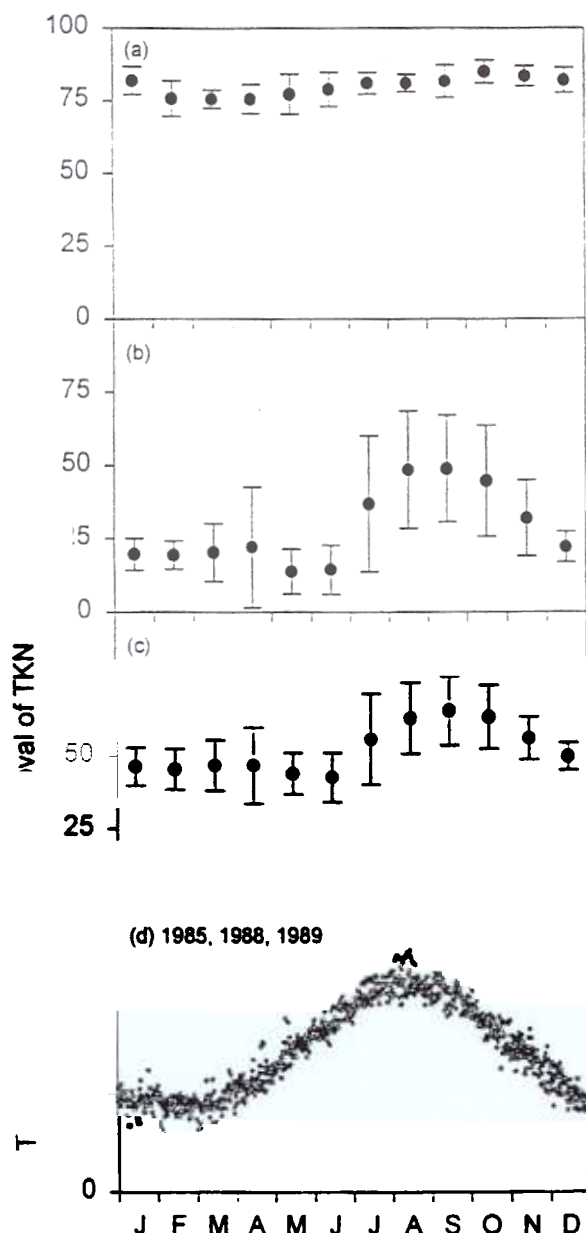


Figure 3—Monthly average ( $\pm$  standard deviation) percent removals of forms of N by Metro for the 1989 to 1996 interval: (a) organic N, (b) T-NH<sub>3</sub>, (c) TKN, and (d) time series of Metro effluent temperature for 3 years.

ment (Figures 4c and 4d), they were low compared with earlier years (Figure 5). During this cold interval, as in previous years, loads of T-NH<sub>3</sub> to Metro were largely transmitted to the lake (Figure 4d, no treatment). Thus reductions in loads from BMS over such intervals are reflected in decreases in loads to the lake from Metro of approximately equal magnitude. The increased load from BMS in early May of 1999 was largely transferred to the lake (Figures 4b and 4d) because it proceeded establishment of effective T-NH<sub>3</sub> treatment (Figure 5). Establishment of some nitrification treatment later in the month, as indicated by the disparity in the influent and effluent concentrations (Figure 4c), somewhat reduced the transfer of the upset load to the lake (Figure 4d). As a result of the BMS upset event, the May 1999 T-NH<sub>3</sub> load from Metro was at the mean value observed before pretreatment (Figure 5). Occurrence of an upset event in the August to October interval

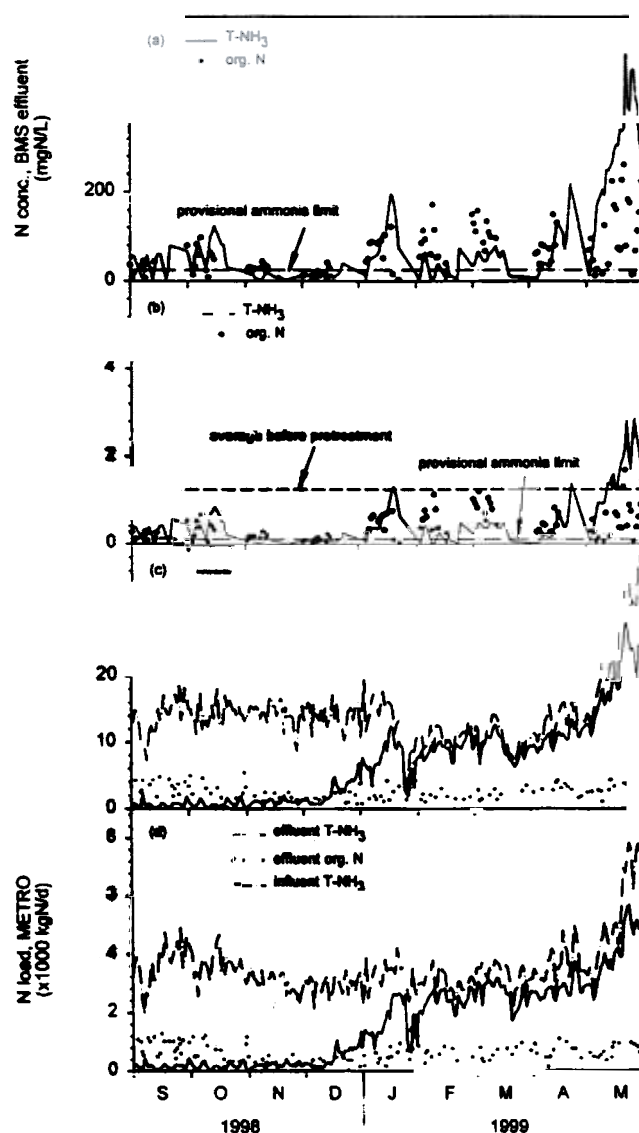
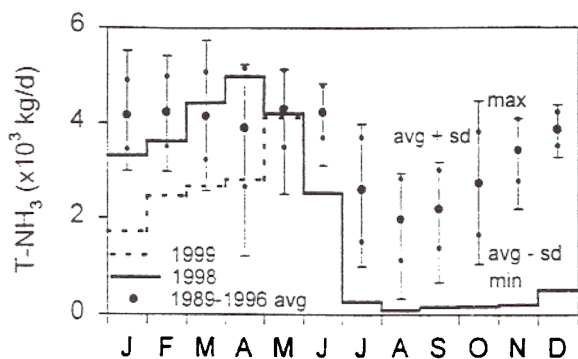


Figure 4—Time series for forms of N over the September 1998 to May 1999 interval: (a) N concentrations in the BMS effluent, (b) N loads from BMS to Metro, (c) N concentrations in influent and effluent of Metro, and (d) N loads to Metro and from Metro to Onondaga Lake.



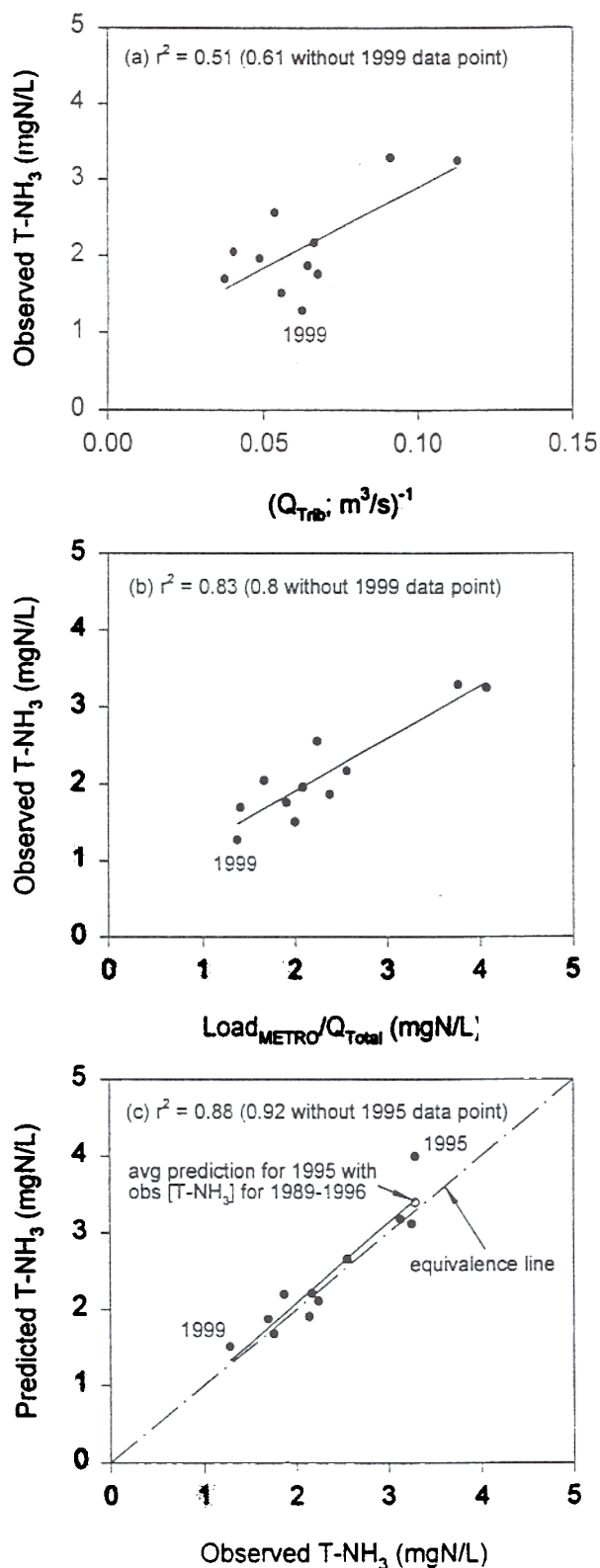
**Figure 5—Comparison of monthly loads from Metro to Onondaga Lake; 1998 and 1999 (January to May) versus observations for the 1989 to 1996 interval (mean  $\pm$  1 standard deviation and the range).**

would not have resulted in such a nearly complete transfer to the lake (Figure 5) because treatment at Metro would have reduced, or perhaps eliminated, this additional load.

Organic N concentrations and loads were typically greater than T-NH<sub>3</sub> levels during the interval of effective T-NH<sub>3</sub> treatment, but remained much lower than this form of N in 1999 (Figures 4c and 4d). The more temporally uniform character of organic N in the Metro effluent is consistent with earlier observations (Figure 3a).

**Lake Total Ammonia Concentration at Spring Turnover.** Earlier studies found that the T-NH<sub>3</sub> concentration in the lake at spring turnover (April) could be predicted reasonably well with an empirical expression that only accommodated antecedent tributary flow (Effler et al., 1996a, and Matthews et al., 2000). This relationship was developed in a dilution model format (Effler et al., 1996b, and Manczak and Florczyk, 1971); for example, lake T-NH<sub>3</sub> concentration as a function of the inverse of the summed tributary flow for a preceding interval of specified duration. Such relationships have most often been reported for fluvial systems where a relatively uniform loading source is diluted according to natural variations in runoff (Manczak and Florczyk, 1974). The empirical relationship for the lake is updated in Figure 6a to include 12 years of observations and modified to accommodate tributary flows for a shorter antecedent interval (January to March, instead of December to March). Excluding the 1999 observation, the relationship explains approximately 60% of the interannual variations in the T-NH<sub>3</sub> concentration observed at spring turnover. Thus, the hypothesis (Effler et al., 1996a, and Matthews et al., 2000) that the wide variations in concentration observed among years at this time ( $\sim 1.5$  to  $3.8$  mg N/L) have been in large part caused by varying dilution associated with natural variations in runoff, under relatively constant loading conditions from Metro, is supported. The 1999 concentration deviates the most from the population of observations; the value was approximately 1 mg N/L lower than predicted for that year (Figure 6a). This is consistent with the reduction in antecedent loading from Metro over the January to March interval of 1999 relative to the earlier years. Inclusion of the 1999 observation shifts the best-fit relationship and degrades the overall performance of this empirical model (for correlation coefficient  $r$ ,  $r^2 = 0.51$ , Figure 6a).

Inclusion of the effect of the Metro T-NH<sub>3</sub> load, in addition to that of tributary flow, represented as the ratio of the load to the flow, results in a stronger empirical model for the spring turnover



**Figure 6—Evaluation of model performance for predicting the concentration of T-NH<sub>3</sub> at spring turnover (April) in Onondaga Lake: (a) empirical model based on tributary flow, (b) empirical model based on tributary flow and Metro load, and (c) mechanistic N model.**

T-NH<sub>3</sub> concentration for the 1988 to 1998 interval ( $r^2 = 0.80$ , Figure 6b). Further, the T-NH<sub>3</sub> observation for 1999 is not an outlier according to this model ( $r^2 = 0.83$  for the 1988 to 1999 interval, Figure 6b), but is predictable from the distinctly lower loading from Metro in that year. This model is inherently more mechanistic (a steady-state representation) than one based strictly on tributary flow; that is, the ratio of the load and flow has units of concentration (x-axis of Figure 6b). Addition of the Metro flow and tributary T-NH<sub>3</sub> loads does not result in a markedly stronger empirical model.

The population of tributary flows for the 12 years included in the above empirical models was not uniformly distributed (Figures 6a and 6b) because there is an absence of intermediate flows within the 12 years, conditions that are not well represented in the data base for the complete 27 year period of record (Gelda et al., 2001). Three of the four lowest flow years for the January to March interval for the 27-year period of record were included in the 1988 to 1999 period. Thus, the effects of low-flow (critical) conditions were well represented in the above empirical models.

The mass-balance N model performed well in predicting the spring turnover T-NH<sub>3</sub> concentrations ( $r^2 = 0.88$ ) and better than the empirical models (Figure 6). This supports the veracity of the mass-balance N model for simulating T-NH<sub>3</sub> concentrations at spring turnover, driven only by detailed hydrology and material loading information from Metro. The high performance of the model establishes that the low T-NH<sub>3</sub> concentration in the lake in the spring of 1999 was a result of reduced antecedent loads from Metro. Further, the position that T-NH<sub>3</sub> behaves in an essentially conservative manner over the cold late fall-to-spring interval is supported (Gelda et al., 2001, and Matthews et al., 2000). This is somewhat fortuitous because there are only limited lake data during this interval (ice cover) to support model testing. The greatest deviation from observations was predicted for 1995, the year of the greatest Metro effluent concentrations (4.4 mg N/L higher than the average for 1988 to 1996). Use of the average Metro effluent T-NH<sub>3</sub> concentrations reported for the 1988 to 1996 interval with the other conditions of 1995 resulted in a prediction that better matched the observations (Figure 6c). This, together with a lack of explanation for the greater effluent concentrations in that year, suggests that the effluent concentrations for the antecedent interval of 1995 may be falsely high.

The mass-balance model represents a credible quantitative framework to resolve the relative contributions of increased treatment capacity at Metro versus pretreatment at BMS in achieving the reductions in T-NH<sub>3</sub> concentrations observed at spring turnover in 1999. All simulations conducted to support this analysis were initialized according to the last observations made in the fall of 1998. The overall benefit emerges by comparison of the observed turnover concentrations of 1999 (~ 1.3 mg N/L) with those predicted by the model (an average of 2.4 mg N/L) by adopting historic Metro effluent concentrations (Figure 7). The range of predictions for historic conditions (1989 to 1996, exclusive of 1995) was rather small, approximately 0.4 mg/L (Figure 7). Thus, the combined (two) rehabilitative efforts reduced the spring turnover concentrations by more than 1 mg N/L (1.1 mg N/L, on average). The effect of pretreatment at BMS was bracketed by two simulations: (1) by adding a T-NH<sub>3</sub> load to the observed Metro effluent load over the fall (October) 1998 through spring 1999 interval that results in inputs that correspond to the estimated average load from this industry before construction of the pretreatment facility and (2) by adding this same BMS load, but not

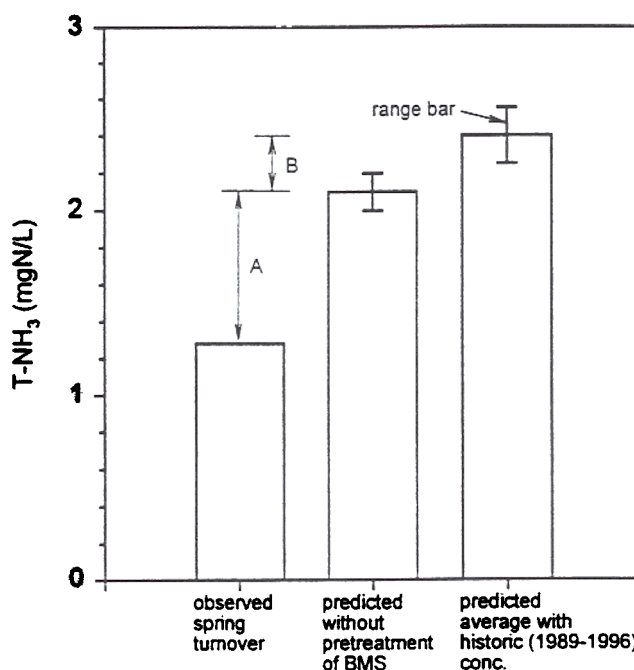
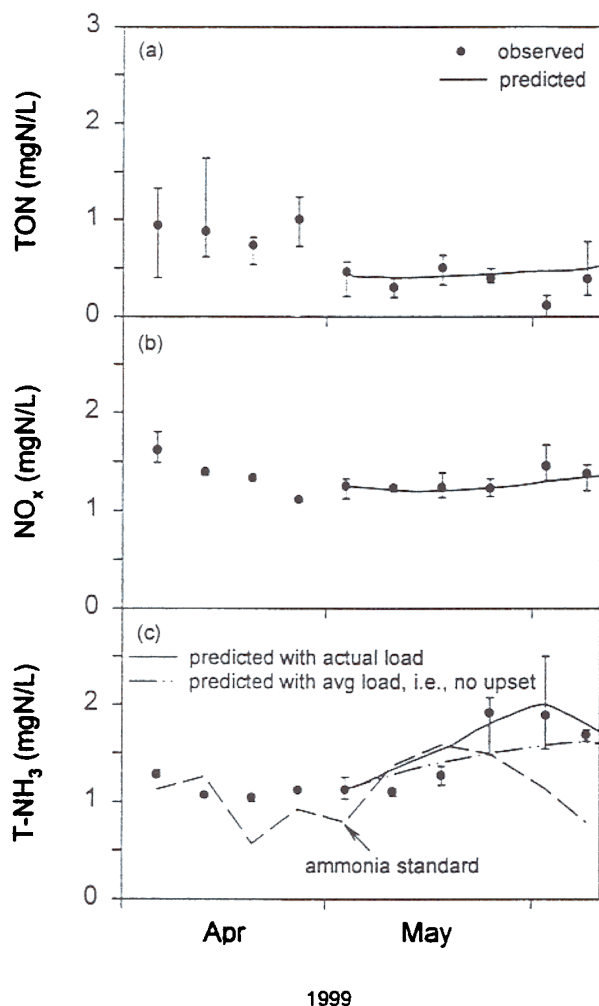


Figure 7—Evaluation of the relative contributions of increased treatment capacity at Metro versus pretreatment at BMS in reducing the T-NH<sub>3</sub> concentrations in Onondaga Lake at spring turnover in 1999, using the N mass-balance model. The magnitudes of A and B correspond to the concentration reductions associated with the pretreatment and increased Metro treatment, respectively. The dimensions of the vertical bars for the two model scenarios correspond to reasonable ranges for the predictions.

starting until January of 1999. The second scenario assumes that the BMS load that prevailed before commencement of pretreatment would have been completely treated through December (Figure 4c), because of Metro's increased treatment capability. The range of predictions associated with these bracketing runs is modest (~ 0.2 mg N/L). The mean (~ 2.1 mg N/L) is considered to be a fair representation of the concentration that would have occurred as a result of increased treatment at Metro in the absence of pretreatment at BMS (Figure 7). Accordingly, approximately 75% of the reduction in T-NH<sub>3</sub> concentration observed at spring turnover in 1999 was attributable to pretreatment at BMS, the remainder was associated with increased treatment capacity at Metro (Figure 7). The dominance of the pretreatment influence is a manifestation of the lake's high flushing rate. The lake experienced approximately 1.15 flushes (on a completely mixed basis) over the January to March interval of 1999, when little ammonia treatment was achieved. Accordingly, the lake concentration at spring turnover was largely regulated by the greater Metro loads over that rather short interval because the lake reached approximately 68% of steady state with that loading level in response to the rapid flushing. Thus the reductions in loading from BMS over that interval had important implications for the lake.

**Temporal Patterns in the Lake in 1999.** There was only modest temporal structure in the concentration of NO<sub>x</sub> in the UML of the lake over the April to early June interval of 1999. The concentration decreased progressively through April from approx-



**Figure 8—Temporal patterns in forms of N in the upper waters of Onondaga Lake in 1999, with simulations from a mass-balance model for a portion of the interval: (a) total organic nitrogen (TON), (b)  $\text{NO}_x$ , and (c)  $\text{T-NH}_3$ , with two simulations and the pattern for the CCC. Vertical bars represent the range of concentrations within the upper layers.**

imately 1.6 mg N/L to approximately 1.1 mg N/L, remained largely unchanged through May, and increased by approximately 0.3 mg N/L in early June (Figure 8b). Only minor vertical differences were observed within the UML. Organic N concentrations averaged approximately 1 mg N/L in April and approximately 0.4 mg N/L in May (Figure 8a). At least part of this decrease was associated with the loss of most of the phytoplankton component during the same interval (deposition; Canale et al., 1996). The strongest phytoplankton bloom occurs in April in most years, followed by a minimum in May (Effler, 1996), that is driven by zooplankton grazing (Siegfried et al., 1996). The most conspicuous structure for forms of N was the abrupt increase in  $\text{T-NH}_3$  in late May. The approximately 0.7 mg N/L increase in  $\text{T-NH}_3$  over 1 week (Figure 8c) was greater than observed in weekly monitoring over the previous 11 years (Brooks and Effler, 1990, and Matthews et al., 2000). The timing of this abrupt increase coincides with the peak in the  $\text{T-NH}_3$  load discharged from Metro (Figure 4d), driven

by the peak in loading from the industrial pretreatment facility (Figure 4b). Variations in the ammonia standard over the study interval reflect the dependence of this limit on ambient pH and T (U.S. EPA, 1985). Concentrations of  $\text{T-NH}_3$  were close to the standard in 1999 through mid-May (Figure 8c); this limit was not exceeded on three of seven monitoring dates over this interval, a substantial improvement compared with conditions documented over the 1988 to 1998 period (Matthews et al., 2000). The observations of late May and early June exceeded the standard by a substantial margin (Figure 8c).

The modeling analysis conducted to delineate the effect of the upset event on the lake  $\text{T-NH}_3$  concentrations and status with respect to the standard (Figure 8c) was based on two simulations. Each were initialized on the first sampling day in May, at the beginning of the pretreatment upset event (Figure 4). The first simulation adopts the estimated dynamics of the Metro load (Figure 4d). The second simulation projects concentrations that would have been observed if the upset event had not occurred; the average Metro load for the March to April interval (Figure 4d) was adopted for this case. The difference in these projections is a fair representation of the effect of the upset event (Figure 8c). The observed increase in the  $\text{T-NH}_3$  concentration and the levels of TON and  $\text{NO}_x$  were well simulated (Figure 8). The concentration of  $\text{T-NH}_3$  would have remained substantially lower in early June (by 0.5 mg N/L) if the upset event at the pretreatment facility had not occurred; accordingly, the standard may not have been violated for the last sampling in May (Figure 8c).

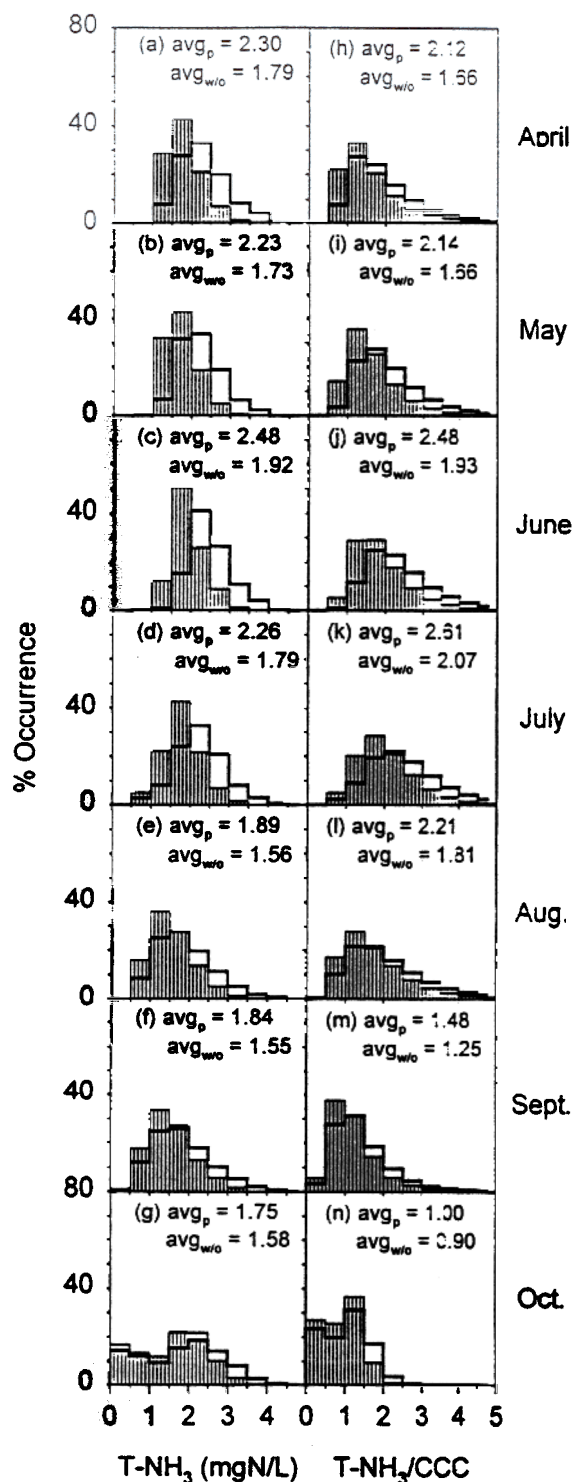
**Projections of Effects of Pharmaceutical Ammonia Loads.** The effect that loads from the pharmaceutical facility had on  $\text{T-NH}_3$  concentrations and violations of the chronic standard in Onondaga Lake is depicted here through comparison of predicted monthly frequency distributions (Figure 9) for conditions that prevailed before establishment of pretreatment and the most recent upgrade at Metro (aeration system) for  $\text{T-NH}_3$  treatment versus the case of suppressing the industrial contribution. The Metro  $\text{T-NH}_3$  load was reduced by 30% for the second case (Tables 1 and 2). The prevailing conditions, and associated inputs to the probabilistic framework, were documented by Gelda et al. (2001). The monthly distributions were based on 81 000 ( $2700 \times 30$ ) daily simulations by the model (Gelda et al., 2001). These simulations indicate that lake concentrations would have been from approximately 10 (October, Figure 9g) to more than 20% (April, Figure 9a) lower in the absence of the  $\text{T-NH}_3$  load from the pharmaceutical facility. Thus, this industrial source exacerbated the lake's ammonia problem by increasing  $\text{T-NH}_3$  concentrations and increasing occurrence and margin of violations of the receiving water standard (Figure 9). Violations would have occurred on 5 to 12% fewer days in the upper waters of the lake in the absence of the pharmaceutical facility's contribution to the Metro load (Figures 9h to 9n).

The potential relative effects of the occurrence of upsets at the pretreatment facility in the future, with respect to meeting water quality goals, are illustrated by application of the probabilistic framework for three selected scenarios (Table 3). The base case (scenario 1, Table 3) adopts the level of nitrification treatment specified for phase 3 in the existing rehabilitation plan (June to October, 1.0 mg N/L; November to May, 2.0 mg N/L). Other inputs (e.g., Metro flows, lake pH, T, etc.) for all three scenarios were as specified by Gelda et al. (2001). Specification of loading from Metro for the pretreatment upset scenarios is necessarily speculative. These events have been assumed to be of the same magnitude (i.e., load and duration) as documented here for May

1999, and to occur randomly within a year, once every year. Two event cases are considered. In the first case (scenario 2, Table 3), Metro is assumed to be 50% effective in treating the additional T-NH<sub>3</sub> load from the pharmaceutical facility. The entire increase in load to Metro associated with the event is assumed to be transmitted to the lake in the second case (scenario 3, Table 3). The results are presented as counts of violations—exceedances of the standard or criterion. According to U.S. EPA guidance documents

**Table 3—Projection of status with respect to chronic ammonia standard—criteria for selected pretreatment upset scenarios.**

Metro loading scenarios for pretreatment upsets	Violation/Exceedance Counts			
	U.S. EPA, 1985		U.S. EPA, 1998	
			4-day	30-day
(1) No increase in loading from Metro.	194	10	25	
(2) 50% of pretreatment upset load to lake.	236	15	32	
(3) 100% of pretreatment upset load to lake.	284	22	48	2



(U.S. EPA, 1998 and 1999), the criterion should not be exceeded more than once every 3 years. Within the context of the probabilistic framework that includes 27 years of simulations (Gelda et al., 2001), this corresponds to not exceeding a total of 9 counts. Projections are presented for two different criteria (one [U.S. EPA, 1985] coincides numerically with the existing state standard) and two different intervals for averaging the lake's status with respect to the limit(s) (Table 3) because these alternatives for evaluating the lake's status are within the prerogatives of the regulatory community.

These projections with the probabilistic framework (Table 3) serve to demonstrate that the continuation of upset events at the pretreatment facility of the pharmaceutical manufacturer could substantially increase the occurrence of violations of an ammonia standard in Onondaga Lake, and thereby compromise the overall rehabilitation plan for municipal wastewater for this system. Wide differences in status are projected depending on the limit and the manner of its application (Table 3). Clearly, the potential importance of these upsets depends on the specifics of the related policy adopted by regulators, including the setting and application of standards (Table 3). Not included in these scenarios is a projection of an upset in nitrification at Metro, that could lead to even greater short-term loads (e.g., domestic plus industrial components). The pharmaceutical facility is also a concern in this regard because of

**Figure 9—Projections with probabilistic framework of T-NH<sub>3</sub> concentrations and status with respect to the New York State chronic ammonia standard for the upper waters of Onondaga Lake. Comparison of monthly distributions for prevailing loads versus the case of elimination of the contribution of the pharmaceutical industry: (a) T-NH<sub>3</sub> concentrations in April, (b) T-NH<sub>3</sub> concentrations in May, (c) T-NH<sub>3</sub> concentrations in June, (d) T-NH<sub>3</sub> concentrations in July, (e) T-NH<sub>3</sub> concentration in August, (f) T-NH<sub>3</sub> concentrations in September, (g) T-NH<sub>3</sub> concentrations in October, (h) T-NH<sub>3</sub>/CCC in April, (i) T-NH<sub>3</sub>/CCC in May, (j) T-NH<sub>3</sub>/CCC in June, (k) T-NH<sub>3</sub>/CCC in July, (l) T-NH<sub>3</sub>/CCC in August, (m) T-NH<sub>3</sub>/CCC in September, and (n) T-NH<sub>3</sub>/CCC in October. Averages of distributions for prevailing conditions (avg<sub>p</sub>) and the case of the absence of pharmaceutical contributions (avg<sub>w/o</sub>) are presented.**

the potential presence of antibiotics and chlorinated solvent (Engineering Science, 1992) in its treated wastewater that could impart toxic effects on treatment microbes at Metro.

### Summary

Loads of TKN ( $\sim 2900$  kg/d) and T-NH<sub>3</sub> from a pharmaceutical manufacturing facility represented approximately one-third of the total loads to Metro before construction of a pretreatment facility for this industrial waste. The T-NH<sub>3</sub> load was transmitted to polluted Onondaga Lake during the colder months, though partial treatment was achieved at Metro in some years during the warmer months. A recently completed pretreatment facility is expected to reduce this industrial load by more than 85%. This is an important component of an expensive larger rehabilitation program for municipal wastewater that is intended to eliminate violations of the ammonia standard in the lake and to meet other water-quality goals.

The responsiveness of the lake to reductions in external loading was demonstrated by the lower T-NH<sub>3</sub> concentrations observed in the upper waters of the lake in the spring of 1999. Analysis with a previously tested mass-balance model, which used detailed loading information from Metro and the pretreatment facility, demonstrated that the lower concentration at spring turnover in 1999 was primarily ( $\sim 75\%$ ) caused by reductions in loading from the pharmaceutical facility achieved by pretreatment and secondarily ( $\sim 25\%$ ) by improved treatment at Metro. An abrupt increase in loading from the pretreatment facility (an upset event) occurred in May 1999 during which loading rates of T-NH<sub>3</sub> corresponded approximately with levels that prevailed before treatment of the industrial waste. This event imparted a conspicuous signature in the lake, an abrupt increase in T-NH<sub>3</sub> concentration that was well simulated with the mass-balance model. The event caused an increase in the T-NH<sub>3</sub> concentration in the upper waters of the lake of approximately 0.5 mg/L, and may have resulted in violations that would not have occurred in the absence of the pretreatment upset.

Application of a probabilistic modeling framework demonstrated that the T-NH<sub>3</sub> load from the pharmaceutical facility, before pretreatment was established, exacerbated the ammonia problem in Onondaga Lake by increasing the occurrence and margin of violation of the chronic toxicity standard in the lake. Projections for speculative scenarios of continued upsets at this facility indicate that such events could compromise the portion of the lake rehabilitation program directed at ammonia. Designs for more advanced biological treatment at Metro should reflect the potential for continued irregular loading inputs from the pretreatment facility.

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